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<p>(54) Title: MULTILAYER PRESSURE-SENSITIVE ADHESIVE CONSTRUCTION</p>		
<p>(57) Abstract</p> <p>The present invention is directed toward a multilayer, pressure-sensitive adhesive construction wherein one layer is formed of a first adhesive composition having a first glass transition temperature, and at least a second layer is formed of a second adhesive composition having a second glass transition temperature. The first glass transition temperature differs from the second glass transition temperature.</p>		

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MULTILAYER PRESSURE-SENSITIVE ADHESIVE CONSTRUCTION

Field of the Invention

The present invention relates to pressure-sensitive adhesive constructions, and more particularly, to multilayer pressure-sensitive adhesive constructions which exhibit both good adhesion and good convertibility.

Background of the Invention

A conventional pressure-sensitive adhesive (PSA) label construction comprises a laminate of a facestock, a pressure-sensitive adhesive layer, and a coated release liner. The facestock may comprise any of a variety of materials, but is typically formed from paper or plastic films. The release liner provides a backing from which the facestock and the pressure-sensitive adhesive are peeled away just prior to label application. The surface of the release liner often consists of paper coated with a release layer of silicone.

Pressure-sensitive adhesive tape and label constructions are usually manufactured as a continuous roll in various widths, and are then processed to form finished product consisting of commercially useful labels or tape rolls. Such processing, known as converting, often involves cutting all or part of the bulk laminate roll. For example, one common converting operation in label manufacture is die cutting and matrix stripping, which involves precision cutting through the facestock and adhesive layers up to but not through the release surface, thereby cutting outlines of the labels, and then pulling away the surrounding matrix to leave only the individual labels on the release liner. Other converting operations may include butt cutting, guillotining, hole punching, slitting, and printing.

The cost of converting the bulk laminate PSA construction into the finished product depends in large part on the speed in which the converting processes can be carried out. The faster the PSA construction can be converted, the lower the cost of the finished product. While most of the current narrow-web converting presses operate at speeds of less than 500 feet per minute, newer modern wide-web converting presses are designed to be operated at speeds of as high as 800 feet per minute or greater, and it is desirable to manufacture PSA constructions compatible with this converting speed.

It has been discovered that all layers of the laminate have some effect on

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converting speed, and much work has been directed at optimizing the facestock and release surfaces for faster converting. For example, increasing matrix stripping speed generally increases stripping force, which often results in matrix breaks which force press shutdown. This problem may be avoided by the use of higher strength facestocks, which convert better than low strength facestocks at a variety of converting speeds.

The adhesive layer, however, has been the greatest limiting factor with respect to the speed of converting bulk laminates into finished product. It is desirable to have an adhesive layer with good flow properties that can adhere to a wide variety of substrates. However, adhesive compositions which are formulated to have these properties do not always convert well, oftentimes sticking to the cutting dies, smearing on the matrix and label edges, and interfering with the precision cutting, or otherwise slowing down the converting process.

In addition, adhesive layers may also impact the matrix stripping operations which follow die cutting, causing breaks in the matrix if the converting press is run at too high a speed. To avoid these matrix breaks, press operators are often forced to slow the converting presses to well below the optimal operating speed.

Thus, it is desirable to provide pressure-sensitive adhesive constructions which feature adhesive layers which show good adhesion to a wide variety of substrates of varying roughness, and which are also compatible with optimal converting performance.

Summary of the Invention

The present invention is directed toward pressure-sensitive adhesive constructions which show good adhesion to a wide variety of substrates, and which also convert well.

In one aspect of the present invention, there is provided a pressure-sensitive adhesive construction with a facestock. A first adhesive layer is on the facestock. The first adhesive layer comprises a first adhesive composition with a first glass transition temperature. The first adhesive composition may be either an acrylic based or a rubber-based adhesive and may include a first organic additive.

A second adhesive layer is on the first adhesive layer. The second adhesive layer comprises a second adhesive composition with a second glass transition

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first storage modulus at a frequency of 10^4 radians per second at 20°C is greater than about 3×10^8 dynes/cm², and the first tangent delta at 10^2 radians per second at 20°C is greater than about 0.5. In this embodiment, the first layer may comprise a pressure-sensitive adhesive layer, or it may comprise an adhesive layer which does not exhibit pressure-sensitive adhesive properties.

Brief Description of the Drawings

FIGURE 1 is a cross-sectional view of a conventional pressure-sensitive adhesive construction.

FIGURE 2 is a cross-sectional view of a pressure-sensitive adhesive construction of the present invention.

FIGURE 3 is a plot of the loss modulus, the storage modulus, and tangent delta of adhesive Formulation 2 in Table I as a function of temperature at 10 radians per second.

FIGURE 4 is a plot of the loss modulus, the storage modulus, and tangent delta of adhesive Formulation 1 in Table I as a function of temperature at 10 radians per second.

FIGURE 5 is a schematic diagram of a dual die used to apply the adhesive formulations of the present invention.

FIGURE 6 is a cross-sectional schematic view showing the lip structure of the dual die of FIGURE 5 as it is applying two layers of adhesive.

FIGURE 7 is a cross-sectional schematic showing an angled lip structure of the dual die as it is applying two layers of adhesive.

FIGURE 8 is a cross-sectional schematic of a dual die featuring a beveled lip structure.

Detailed Description of the Preferred Embodiments

The present invention is directed to multilayer adhesive constructions showing improved convertibility as well as good adhesion and aging.

In one embodiment of the present invention, this is achieved by providing a multilayer pressure-sensitive adhesive construction which comprises two or more adhesive layers, at least one of which is a pressure-sensitive adhesive layer, and where at least one of the layers has an adhesive composition with a glass transition temperature that differs from the glass transition temperature of an adhesive

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composition in a different layer. In this embodiment, it is preferred that the adhesive layer nearest the facstock contain the adhesive composition with the highest glass transition temperature. The adhesive layer nearest the release layer has the adhesive composition with the lower glass transition temperature, and may be selected for its desirable adherent properties. It has been discovered that multilayer adhesive constructions of this design, which feature at least two adhesive layers with differing glass transition temperatures, show markedly improved convertibility relative to conventional pressure-sensitive adhesive constructions.

In another multilayer embodiment of the present invention, the adhesive layer in contact with the facstock has a lower glass transition temperature than an adhesive layer nearer to the release surface. In this aspect of the present invention, the multiple layers of the adhesive construct can be selected and arranged to balance improved converting with desirable adherent properties.

As used herein, the term "glass transition temperature" (T_g) refers to the temperature at which an adhesive composition, which may include polymers, resins, and oils, and other ingredients, changes from a glassy to a rubbery state. For block copolymer based adhesives (SIS, SBS, SI, SB, SEBS, or other block copolymers with endblocks of polystyrene), the T_g reflects only the midblock of the elastomer components of the adhesive composition. The term "native glass transition temperature," as used herein, refers to the glass transition temperature of a particular component of the adhesive composition, such as a polymer, as that component exists in its pure form.

In another embodiment of the present invention, a multilayer pressure-sensitive adhesive construction with improved convertibility and good adhesion is achieved by providing a multilayer adhesive construction with a first layer, wherein the first layer has a storage modulus which, at a frequency of 10^4 radians per second at 20°C , is greater than about 3×10^8 dynes/cm², and a tangent delta, which at 10^2 radians per second and 20°C , is greater than about 0.5. A second adhesive layer comprising a pressure-sensitive adhesive may be in contact with the first layer, thereby providing an adhesive layer which shows good adhesion to a wide variety of substrates. Multilayer adhesive constructions of this design also exhibit good convertibility and good adhesion to a variety of substrates of varying surface

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roughness.

Referring to FIG. 1, for comparison purposes, there is depicted a conventional pressure-sensitive adhesive construction. Construction 10 features a facestock 12, a pressure-sensitive adhesive layer 14 of uniform composition in contact with the facestock 12, and a release layer 16, having a release surface thereon, in contact with the pressure-sensitive adhesive layer 14.

Although the present invention will be described in the context of a construction having a release layer, the present invention is equally applicable in the context of a construction such as for the production of tape in which the release layer is omitted and a release surface is provided on the opposite side of the facestock from the adhesive. It is contemplated that tape constructions may benefit from the advantages of the present invention during various converting operations involving cutting of the bulk tape roll, such as slitting.

In addition, although the present invention may be described in the context of having two pressure-sensitive adhesive layers, it is equally applicable to a multilayer adhesive construction where only the adhesive layer nearest the release layer is a pressure-sensitive adhesive, and wherein the other layers may or may not be an inherently tacky adhesive layer, as described more fully below. Thus, the term "adhesive layer," and "adhesive composition," as used herein, are meant only to refer to layers, or the adhesive compositions in those layers, with at least that minimal degree of tack required to adhere to a particular facestock substrate of an adhesive construction. An "adhesive layer" or "adhesive composition" may include, but does not necessarily include, "pressure-sensitive adhesive layers" or "pressure-sensitive adhesives," which possess substantially greater tack than that minimal degree of tack needed to adhere to the facestock, and which adhere well to a variety of substrates on contact.

Referring to FIG. 2, there is depicted one embodiment of the multilayer pressure-sensitive adhesive construction of the present invention which features two pressure-sensitive adhesive layers. Multilayer pressure-sensitive adhesive construction 20 is comprised of a facestock 22, a first pressure-sensitive adhesive layer 24 in contact with the facestock 22, a second pressure-sensitive adhesive layer 26 in contact with the first pressure-sensitive adhesive layer 24, and a release layer

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28 in contact with the second pressure-sensitive adhesive layer 26. Pressure-sensitive adhesive layers 24 and 26 each have one or more polymerized components, as well as other components discussed below, which combine to form an adhesive composition with at least one glass transition temperature. The glass transition
5 temperatures of the adhesive compositions of the respective adhesive layers differs.

As can be readily seen from comparison of FIGS. 1 and 2, pressure-sensitive adhesive constructions of the present invention differ from conventional pressure-sensitive adhesive constructions, in addition to ways described more fully below, by having two layers 24 and 26. In one embodiment, described above, layers 24 and
10 26 are both formed of inherently tacky pressure-sensitive adhesive compositions, and thus comprise pressure-sensitive adhesive layers. In an alternate embodiment, described in more detail below, layer 24 is formed of a polymeric composition which is not inherently tacky, and does not exhibit pressure-sensitive adhesive properties, thereby forming an adhesive layer 26 which is not a pressure-sensitive
15 adhesive layer.

In addition, although not depicted, it is contemplated that the teachings of the present invention are applicable to create multilayer pressure-sensitive adhesive constructions which feature three or more layers, where one or more layers may comprise a pressure-sensitive adhesive, and which show the improved convertibility
20 and aging of constructions of the present invention.

In all embodiments of the present invention, layers 24 and 26 are comprised of polymeric compositions. Various other organic and inorganic substances may be added to the polymeric compositions as may be desired to alter the properties of the layers. For example, additives may be selected to enhance the adhesive properties
25 of a particular layer, as described more fully below.

The polymeric components of layers 24 and 26 may consist of any of a variety of polymers which are known to be useful in the formation of adhesive compositions, such as natural or synthetic elastomers, or acrylic based adhesive compositions. For example, adhesive layers 24 and 26 may both consist of
30 compositions including elastomeric polymers, or they may both consist of compositions including acrylic polymers. Alternately, either layer 24 or 26 may consist of a composition including an elastomeric polymer, with the other layer

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being a composition including an acrylic polymer. Layers 24 and 26 may be formed with identical polymer components or different polymer components.

In addition, although layer 26 should comprise a pressure-sensitive adhesive, and thus should be formed from an adhesive composition which imparts pressure sensitive properties, layer 24 may or may not be formed of a pressure-sensitive adhesive composition. In the preferred embodiment, layer 24 only need possess sufficient tack to adhere to facestock 22 when laminated at room temperature or elevated temperature. However, as can be appreciated by one of skill in the art, the polymeric compositions in layers 24 and 26 must be sufficiently compatible to adhere to one another strongly enough to prevent separation of layers 24 and 26 when the facestock 22 and layers 24 and 26 are removed from the release layer 28, as for example, when a label construction is peeled from its backing.

In one embodiment, either layer 24 or 26, or both layers 24 and 26, may be formed from an acrylic based polymer. It is contemplated that any acrylic based polymer capable of forming an adhesive layer with sufficient tack to adhere to the facestock 22 may function in the present invention. In addition, with respect to layer 24, and layer 26 in certain embodiments, presently preferred acrylic polymers for the pressure-sensitive adhesive layers include those formed from polymerization of at least one alkyl acrylate monomer containing from about 4 to about 12 carbon atoms in the alkyl group, and present in an amount from about 35-95 % by weight of the polymer or copolymer, as disclosed in U.S. Patent No. 5,264,532 issued to Bernard. Optionally, the acrylic based pressure-sensitive adhesive might be formed from a single polymeric species.

Advantageously, the glass transition temperature of an adhesive layer comprising acrylic polymers can be varied by adjusting the amount of polar, or "hard monomers," in the copolymer, as taught by U.S. Patent No. 5,264,532, incorporated herein by reference. The greater the percentage by weight of hard monomers in an acrylic copolymer, the higher the glass transition temperature. Hard monomers contemplated useful for the present invention include vinyl esters, carboxylic acids, and methacrylates, in concentrations by weight ranging from about zero to about thirty-five percent by weight of the polymer.

In another embodiment, layers 24 and 26 each include at least one

unsaturated elastomeric polymer. The elastomeric polymers used are preferably based on di-block and tri-block copolymers of styrene/butadiene and styrene/isoprene. For example, styrene-butadiene-styrene, styrene-isoprene-styrene, styrene-butadiene, and styrene-isoprene block copolymers, such as the Kraton
5 polymers manufactured and sold by Shell Chemical Company, of Houston, Texas and the Solprene polymers manufactured and sold by Housemex, Inc., located in Houston, Texas, are suitable for use in the instant invention. Multibranched styrene-butadiene and multibranched styrene-isoprene, of the formula $(SB)_x$ and $(SI)_x$, respectively, where x is greater than 2, may also be used.

10 Co-blends of Kraton RP 6419, a styrene-isoprene-styrene block copolymer, and Solprene 1205, a styrene-butadiene block copolymer, have proven to be particularly suited for the polymeric compositions of the respective layers. When a co-blended elastomeric polymer composition is used, more than one glass transition temperature may be observed, as taught by U.S. Patent No. 5,290,842,
15 issued to Sasaki, et al., incorporated herein by reference. In such a case the temperature corresponding to the major peak, as observed on a plot of tangent delta as a function of temperature, is the important glass transition temperature for purposes of the present invention.

The polymeric compositions of the respective layers 24 and 26 may both
20 function as pressure-sensitive adhesives, although this is not required for those layers not in contact with the release layer. However, unsaturated elastomeric polymers normally do not function as pressure-sensitive adhesives by themselves. Pressure sensitive properties are imparted to compositions containing unsaturated elastomeric polymers by the addition of other organic molecules known as tackifiers. Tackifiers
25 are generally hydrocarbon molecules, wood resins, pall resins, and the like, which when present in concentrations ranging from about 40% to about 90% by weight of the total adhesive composition, more preferably from about 45% to about 85% by weight, impart pressure-sensitive adhesive characteristics to the elastomeric polymer adhesive formulation. Compositions containing less than about 40% by weight of
30 tackifier additive do not generally show sufficient "quickstick," or initial adhesion, to function as a pressure-sensitive adhesive, and therefore are not inherently tacky. Compositions with too high a concentration of tackifying additive, on the other

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hand, generally show too little cohesive strength to work properly in most intended use applications of constructions made in accordance with the instant invention.

Tackifier additives are well known in the art, and may be used to increase the glass transition temperature of the unsaturated elastomeric polymer adhesive composition, in proportion to increasing weight concentration of the tackifier. Thus, in addition to their ability to impart pressure-sensitive adhesive properties to the compositions of the instant invention, tackifiers may also be selected to optimize the difference of the glass transition temperatures between the adhesive compositions in different layers. For example, with reference to FIG. 2, the glass transition temperature of the adhesive composition in adhesive layer 24 could be increased by increasing the concentration by weight of a tackifying additive in layer 24. Similarly, the glass transition temperature of the adhesive composition in adhesive layer 26 could be decreased by decreasing the concentration by weight of tackifying additive in layer 26.

Tackifier additives useful for practicing the present invention are those which impart pressure-sensitive adhesive properties to the elastomeric polymer compositions, and which also serve to increase the glass transition temperature of the adhesive compositions. It is contemplated that any tackifier known by those of skill in the art to be compatible with elastomeric polymer compositions may be used with the present embodiment of the invention. One such tackifier, found useful for the Solprene 1205/Kraton RP 6419 blends discussed previously, is Wingtak 10, a synthetic polyterpene resin which is liquid at room temperature, and sold by the Goodyear Tire and Rubber Company of Akron, Ohio. Other suitable tackifying additives may include Escorez 1310 and Escorez 2596, both manufactured by Exxon of Irving, Texas. Of course, as can be appreciated by those of skill in the art, a variety of different tackifying additives may be used to practice the present invention.

In contrast to tackifiers, plasticizers are organic molecules, which form liquids at room temperature, and which are known to decrease the glass transition temperature of an adhesive composition containing elastomeric polymers. Thus, plasticizers may be used in the instant invention, alone or in combination with tackifiers, to alter the glass transition temperature of a particular adhesive

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composition in one of the adhesive layers. One plasticizer found useful for practicing the present invention is Shellflex 371, a naphthenic processing oil available from Shell Oil Company of Houston, Texas. However, any plasticizer known by those of skill in the art to be compatible with elastomeric polymer compositions may be used to practice the present invention.

Many liquid tackifiers or plasticizers which might be used with elastomeric polymers are composed of small organic molecules which are capable of migrating between adjacent adhesive layers. Migration is especially likely to occur if the liquid concentration of tackifiers and plasticizers in one elastomeric polymer adhesive layer greatly exceeds that present in an adjacent elastomeric polymer adhesive layer. Migration of these species is undesirable, as it leads to loss of adhesion over time, otherwise known as poor "aging" of the adhesive layer. To minimize the chances for migration to occur, it is preferred that the weight ratio of the sum of elastomeric polymer to the sum of liquid tackifiers and plasticizers in a particular layer, defined herein as the "polymer to liquid tackifier and plasticizer ratio," be roughly equal between adjacent layers.

In addition to tackifiers and plasticizers, other additives may be used in the adhesive compositions to impart desired properties. For example, anti-oxidants may be used to protect the polymers from oxidative degradation. Suitable anti-oxidants useful in practicing the present invention include Irgafos 168 and Irganox 565, available from Ciba-Geigy, located in Hawthorne, New York.

Various cutting agents, such as waxes and surfactants, as taught by U.S. Patent No. 5,322,876, issued to Sasaki, may also be added to the adhesive compositions of the present invention. For example, Pluronic F108, a block copolymer surfactant having polyethylene-oxide and polypropylene-oxide blocks, and manufactured by BASF, has been found useful in practicing the present invention. Polyethylene glycol waxes, such as Carbowax 1450 manufactured by Union Carbide, located in Danbury, Connecticut, may also be used.

Filler components such as Camel Wite, which consists of calcium carbonate, and is available from Genstar Stone Products Co., located in Hunt Valley, Missouri, may also be used in the adhesive compositions of the present invention.

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Representative formulations of adhesive compositions useful in practicing the present invention are set forth in Table I. All values listed are parts by weight.

TABLE I

Raw Materials	Formulation (ppw)								
	F1	F2	F3	F4	F5	F6	F7	F8	F9
Kraton RP 6419	20	13							
Kraton 1112				13					
Kraton 1107			23.2		13	16.5	20	23.5	27
Solprene 1205	10	7	10	7	7	8.5	10	11.5	13.0
Escorez 2596	46	64	43.3	64	64	55	46	37	28
Wingtack 10	12	8	23.5	8	8	10	12	14	16
Shellflex 371	12	8		8	8	10	12	14	16
Camel Wite	8.5	8.5	3	8.5	8.5	8.5	8.5	8.5	8.5
Irgafos 168	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Irganox 565	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Pluronic F108			3						
Carbowax 1450			3						

Referring to FIG. 2, it is important to the functioning of the present invention that the glass transition temperatures of the adhesive compositions in adhesive layers 24 and 26 differ. Preferably, the glass transition temperatures should differ by about ten to about fifty degrees Celsius. More preferably, the glass transition temperatures should differ by about ten to about thirty degrees Celsius. In a most preferred embodiment, the glass transition temperatures differ by about fifteen to about twenty-five degrees Celsius.

In one preferred embodiment, the adhesive layer containing the adhesive composition with the higher glass transition temperature is in contact with the facestock 22, and the adhesive composition with the lower glass transition temperature is in contact with the release layer 28. It has been discovered that pressure-sensitive adhesive constructions embodying this design show improved converting properties, as will be discussed below. However, it should be

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appreciated by those of skill in the art that some improved converting embodiments of the present invention may feature the adhesive composition nearest the release layer 28 having a higher glass transition temperature than the layer nearest the facestock 22.

5 For purposes of the present invention, it is the difference between the glass transition temperatures of the adhesive compositions in the adhesive layers that appears to reflect one characteristic important to the functioning of the present invention. In an optimal embodiment of the present invention, the difference in the glass transition temperature between the two layers is such that the layer in contact
10 with the facestock has at least a sufficient amount of tack to adhere to the facestock, while the adhesive layer nearest the release layer is inherently tacky. The degree of tack possessed by a particular adhesive layer is dependent in large part on the glass transition temperature of the adhesive composition of that layer. If the glass transition temperature is too high, the composition will fail to function as an
15 adhesive. In contrast, if the glass transition temperature is too low, the adhesive composition may flow too readily, which diminishes converting performance. Various components of the adhesive composition have native glass transition temperatures, which contribute to the observed glass transition temperature of the adhesive composition as a whole. These components include the polymers,
20 tackifiers, plasticizers, fillers, and various other additives known to those of skill in the art.

 In addition, if a particular adhesive is to function as a pressure-sensitive adhesive, as is required for the adhesive layer nearest the release surface, the adhesive composition should have a glass transition temperature of at least about 5
25 degrees to about 70 degrees Celsius below the contemplated use application temperature, more preferably from about 10 degrees to about 50 degrees below the contemplated use application temperature.

 Another important characteristic to the functioning of the present invention is the storage modulus (G'), which is a measure of the energy stored and recovered
30 in a viscoelastic adhesive composition. Adhesive compositions with a sufficiently high storage modulus for a given amount of force exhibit less deformation, and are therefore less likely to adhere to cutting blades or dies used in the converting

process. In one embodiment of the present invention, it is preferred that the layer with the higher storage modulus be in contact with the facestock 22. The layer with the lower storage modulus is in contact with the release layer 28. In this embodiment it is preferred that the layer in contact with the facestock have a storage modulus, at 10^4 radians per second and 20°C , within the range of from about 1×10^8 dynes/cm² to about 5×10^9 dynes/cm², more preferably in the range from about 3×10^8 dynes/cm² to about 1×10^9 dynes/cm², and most preferably within the range from about 3×10^8 dynes/cm² to about 8×10^8 dynes/cm².

The glass transition temperature and storage modulus of adhesive compositions used in the present invention can be determined by various methods known to those of skill in the art. Because of the viscoelastic nature of the adhesive compositions, the T_g value will be dependent on the type and rate of these methods. One such method is to plot the tangent delta, which is the ratio of the loss modulus (G'') to the storage modulus (G'), as a function of temperature. The temperature at which the tangent delta peak occurs, at a frequency of 10 radians per second, represents the glass transition temperature. The loss modulus (G'') is a measure of the energy dissipated as heat or sound per cycle of sinusoidal deformation, when different systems are compared at the same strain amplitude. In addition, for pressure-sensitive adhesives, loss modulus can be correlated to the amount of energy dissipated in peeling the viscoelastic polymeric material from a substrate.

Tangent delta ($\tan \delta$) is measured by placing an approximately 1.5-2 mm thick sample of an adhesive composition between two 8 mm parallel plates of a Rheometrics instrument (model RMS-800 manufactured and sold by Rheometrics, Inc., Piscataway, New Jersey), and oscillating the plates relative to one another at 10 radians per second. The parallel plates are heated at a rate of 1°C per minute during the test. Measurements of the storage modulus, loss modulus, and tangent delta are made at 3°C intervals. In addition, either the loss modulus, or storage modulus, may be measured individually using a similar protocol, but different frequencies, as is known by those of skill in the art.

FIG. 3 is a plot of tangent delta as a function of temperature for Formulation 2 as disclosed in Table I. The glass transition temperature corresponds to the temperature at which the tangent delta peak occurs at 10 radians per second, and is

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approximately 34 degrees Celsius.

FIG. 4 is a plot of the tangent delta as a function of temperature for Formulation 1 as disclosed in Table I. The glass transition temperature for this composition correspond to the temperature at which the tangent delta peak occurs at 10 radians per second, and is about 10.7 degrees Celsius.

The glass transition temperatures of Formulations 1, 2, and 5-9 of Table I, as well as the storage modulus and tangent delta at 20°C and at various frequencies, as determined using the Rheometrics instrument as discussed above, are set forth below in Table II.

TABLE II

	G' 10 ² radians/s dynes/cm ²	tan δ 10 ² radians/s	G' 10 ² radians/s dynes/cm ²	tan δ 10 ⁴ radians/s	G' 10 ² radians/s dynes/cm ²	tan δ 10 ⁴ radians/s	T _g °C
Formulation 1	2.7 × 10 ⁶	1.8	7.3 × 10 ⁷	0.9	2.3 × 10 ⁶	0.58	10.7
Formulation 2	4.5 × 10 ⁷	1.5	4.2 × 10 ⁶	0.5	7.9 × 10 ⁶	0.35	34
Formulation 5	6.7 × 10 ⁷	1.1	5.0 × 10 ⁶	0.5	8.8 × 10 ⁶	0.3	33.4
Formulation 6	1.0 × 10 ⁷	1.7	1.8 × 10 ⁶	0.7	4.3 × 10 ⁶	0.5	24.5
Formulation 7	1.8 × 10 ⁶	1.7	7.0 × 10 ⁷	1.0	2.3 × 10 ⁶	0.7	10.85
Formulation 8	1.0 × 10 ⁶	0.8	1.6 × 10 ⁶	1.0	4.5 × 10 ⁶	0.7	-0.6
Formulation 9	9.8 × 10 ³	0.8	1.3 × 10 ⁷	1.5	5.0 × 10 ⁷	1.5	-15.5

Facestock 22, as depicted in FIG. 2, may comprise any of a variety of materials known by those of skill in the art to be suitable as a facestock material. For example, facestock 22 may be composed of such materials as paper, polyester, or other polymeric materials suitable for facestock use such as polyethylene or polypropylene. The only requirement for facestock 22 is that it be capable of forming some degree of adhesive bond to adhesive layer 24, preferably by having layer 24 adhere to the material selected as the facestock.

Similarly, release layer 28 may consist of any of a variety of materials known to those of skill in the art. In one preferred embodiment, useful for label manufacture, release layer 28 comprises a silicone coated paper substance.

The thickness of the adhesive layers 24 and 26 is typically characterized in terms of the number of grams of adhesive composition applied per meter squared of the surface it is applied on. Generally, a coating weight of about 25 g/m² is

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roughly equal to a thickness of about 1/1000 of an inch, although this may vary considerably depending on the density and type of adhesive used.

The present invention will work when the individual coat weight in each of adhesive layer 24 and 26 is enough to form a discrete identifiable layer. The total
5 coat weight, defined as the sum of the coat weights of adhesive layers 24 and 26, may vary from 15 g/m² to 125 g/m².

In single layer adhesive constructions using a rubber-based adhesive layer, it has been discovered that convertability decreases in proportion to increasing coat weight of the adhesive layer. In these rubber-based systems, convertability
10 decreases dramatically when the adhesive layer is coated at a weight of greater than 25 g/m². Therefore, in the preferred embodiment of the present invention, the sum of the two coat weights of adhesive layers 24 and 26 ranges from about 15 g/m² to less than about 30 g/m², more preferably to less than about 25 g/m², and most preferably to about 20 g/m².

15 Within the most preferred total coat weight of about 20 g/m², the respective coat weights of individual adhesive layers 24 and 26 may vary considerably. For example, the adhesive layer 24 nearest the facestock may have a coat weight range of from about 5 g/m² to about 15 g/m². Similarly, the adhesive layer 26 furthest from the facestock may also have a coat weight range of from about 5 g/m² to about
20 15 g/m². Set forth in Table III are Examples 1-23, which utilize the adhesive Formulations disclosed in Table I, and demonstrate a variety of coat weight ranges that the individual layers might comprise in nonlimiting exemplary embodiments of the present invention. As used in Table III, "Upper Adhesive Layer" refers to the layer nearest the facestock and "Lower Adhesive Layer" refers to the layer nearest
25 the release liner.

Set forth in Table IV are adhesive data for Examples 1-23 of Table III. Looptack was determined by forming an 8" loop of a 1" wide sample, mounting the loop in the jaws of an Instron tester, and then moving the loop against a test surface at 12" per minute, and after a 1" x 3" area was covered, removing the loop at 12"
30 per minute. The force recorded was reported as looptack. Table IVa discloses looptack data for the selected constructions of Table III at low temperatures on high density polyethylene (HDPE).

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This data demonstrates that multilayer constructions of nonlimiting exemplary embodiments of the present invention show good adhesion to a variety of substrates, and also show good adhesion at low temperature. In addition, although not shown, it has also been discovered that similar multilayer adhesive constructions do not lose
5 significant adhesive strength on aging at room temperature for up to three months.

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TABLE III

	Facestock	Upper Adhesive Layer	Upper Layer Coat Weight	Lower Adhesive Layer	Lower Layer Coat Weight	Release Layer
Example 1	HG	F1	15 g/m ²	F2	5 g/m ²	BG
Example 2	HG	F2	5 g/m ²	F1	15 g/m ²	BG
Example 3	HG	F2	10 g/m ²	F1	10 g/m ²	BG
Example 4	HG	F2	15 g/m ²	F1	5 g/m ²	BG
Example 5	HG	F1	20 g/m ²	--	--	BG
Example 6	HG	F2	20 g/m ²	--	--	BG
Example 7	Polyester	F2	5 g/m ²	F1	15 g/m ²	BG
Example 8	Polyester	F2	10 g/m ²	F1	10 g/m ²	BG
Example 9	Polyester	F2	15 g/m ²	F1	5 g/m ²	BG
Example 10	Polyester	F1	20 g/m ²	--	--	BG
Example 11	HG	F5	10	F6	10	BG
Example 12	HG	F5	10	F7	10	BG
Example 13	HG	F4	10	F8	10	BG
Example 14	HG	F4	10	F9	10	BG
Example 15	HG	F6	10	F6	10	BG
Example 16	HG	F6	10	F7	10	BG
Example 17	HG	F6	10	F8	10	BG
Example 18	HG	F7	10	F7	10	BG
Example 19	HG	F7	10	F8	10	BG
Example 20	HG	F5	20	F7	0	BG
Example 21	HG	F5	15	F7	5	BG
Example 22	HG	F5	5	F7	15	BG
Example 23	HG	F5	0	F7	20	BG

HG = High Gloss white paper

F1 = Formulation 1 from Table I

F2 = Formulation 2 from Table I

BG = Silicon coated 40 lb. backing paper

Polyester facestocks used were composed of MYLAR, manufactured by DuPont Chemical Co. of Wilmington, Delaware

TABLE IV

	Stainless Steel	Cardboard	High Density Polyethylene
Adhesive Const. from Table III	Average Initial Looptack N/m	Average Initial Looptack N/m	Average Initial Looptack N/m
Ex. 1	1090 (± 110)	177 (± 59)	760 (± 200)
Ex. 2	730 (± 120)	224 (± 50)	587 (± 40)
Ex. 3	800 (± 200)	152 (± 44)	585 (± 37)
Ex. 4	690 (± 150)	75 (± 9)	450 (± 110)
Ex. 5	650 (± 46)	147 (± 35)	420 (± 120)
Ex. 6	168 (± 43)	1.8 (± 16)	111 (± 32)
Ex. 7	720 (± 68)	265 (± 68)	330 (± 170)
Ex. 8	776 (± 26)	182 (± 32)	301 (± 78)
Ex. 9	615 (± 33)	100 (± 25)	98 (± 33)
Ex. 10	648 (± 21)	305 (± 20)	350 (± 110)
Ex. 11	460	550	117
Ex. 12	555	780	241
Ex. 13	640	750	264
Ex. 14	520	840	266
Ex. 15	690	740	145
Ex. 16	690	849	317
Ex. 17	613	918	249
Ex. 18	508	805	254
Ex. 19	454	682	239
Ex. 20	45	570	18
Ex. 21	370	770	103
Ex. 22	649	740	237
Ex. 23	508	805	254

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TABLE IVa.

Adhesive Construction From Table III	Looptack at 0°C on HDPE N/m	Looptack at 5°C on HDPE N/m
Ex. 11	2	
Ex. 12	19	
Ex. 13	64	11
Ex. 14	344	98
Ex. 15	2	
Ex. 16	36	
Ex. 17	114	26
Ex. 18	167	32
Ex. 19	430	183
Ex. 20	1.2	
Ex. 21	1.4	
Ex. 22	35	2
Ex. 23	167	

The adhesive layers of the present invention may be coated onto facestock or release liners by any means known to those of skill in the art. For example, it is contemplated that adhesive layers 24 and 26 may be applied by solvent coating, hot melt coating, or emulsion coating, at one or more coating stations. Adhesive layers 24 and 26 might also be coated to different substrates, and then be laminated together to form an integral product. Other known methods of simultaneous coating include slide coating, multilayer die coating, or die/slide combination coating.

One preferred method of manufacture uses a multilayer die 50 such as that

illustrated in FIG. 5. Although the die shown in FIG. 5 illustrates the application of two coating layers to a substrate 52, it will be understood that the principles of this method are equally applicable to a plurality of layers in addition to two. In accordance with standard practice, the substrate, which in this case preferably comprises silicone coated paper, is referred to as a "web" and is formed into a long roll. The web 52 travels around a back-up roll 54 as it passes the distal end of the multilayer die 50. As shown in FIG. 5, it will be understood that both the die 50 and the web 52 have substantially equal width such that the entire width of the substrate is coated in one pass by the fluid flowing out of the die 50 and onto the web 52. In this case, two separate fluid layers are flowing out of manifolds 56 formed in the die and along individual slots 60 which are defined by the die's distal lands 62. The slots 50 each communicate with the interface between the web 52 and the distal most tips 58 of the die 50. These tips are referred to as the "die lips" 58 and are illustrated and described in more detail in connection with FIG. 6 below.

The multilayer die 50 is modular, thus allowing for variations in the individual slots 60 and lip 58 configurations without necessitating modifications to the other slots and lips. Thus, these geometries can be adjusted in order to achieve successful coating. Other variables include the "coating gap" (e.g.) and the "angle of attack" (a) of the die. As illustrated in FIG. 5, the coating gap is the distance that the lips 58 are set back from the web. The angle of attack (a) is the degree of angular adjustment of the lip surfaces and of the entire die with respect to the outer pointing normal of the web as illustrated in FIG. 7. Another variable is the web speed which may vary between 50-1,000 feet per minute, and more.

Either one of two die coating methods may be utilized: interference coating or proximity coating. In the former case, the lips 58 of the die actually are pressed forward in the direction of the web 52, but do not contact the web nor cause any damage thereto, because they hydroplane on a thin layer of coating material. However, the pressure may actually cause the back-up roll 54 (typically constructed from a hard rubber material) to deform in order to relieve the pressure of the die against the lips 58. In proximity coating, the lips 58 of the die 50 are positioned a precise distance from the web 52 and are not pressed forward toward the web. The back-up roll 54 is typically constructed from a stainless steel material which allows

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for precision in the circumference of the roll and minimizes roll run-out. The method described herein can be successfully utilized with either type of coating technique.

5 Thus, since very thin layers of high viscosity adhesives are being coated at relatively high web speeds, the process must be carefully controlled.

Such control is accomplished with the present multilayer die coating technique, in part due to the geometry and configuration of the die lips 58. Referring to FIG. 6, there is shown a close-up view of the distal most tips 62 of the multilayer die of FIG. 5, including the lips 58 associated with each slot, showing the
10 interface or coating gap with respect to the web 52. With respect to FIG. 6, it should be noted that, for ease of illustration, the die 50 is shown rotated 90° from the position shown in FIG. 5. Moreover, the web 52 is shown in a horizontal arrangement, when in actuality, there may be a slight curvature to the web 52 and back-up roll (not shown) at this point; however, the distances involved are so short
15 that a good approximation of the fluid dynamics can be achieved by assuming a horizontal web 52.

For consistent reference, the individual lips 58 of the multilayer die 50 shall be referred to with respect to the direction of travel of the web 52. For example, the lip 58a shown to the left in FIG. 6 will be referred to as the "upstream lip,"
20 while the right-most lip 58c shall be referred to as the "downstream lip." Thus, the "middle lip" 58b will have that same reference. Accordingly, the upstream and middle lips 58a, 58b define an upstream feed gap 64 through which an adhesive material 66 flows onto the web 52 to form a bottom adhesive layer 68 of a multilayer adhesive product. Likewise, the middle lip 58b and the downstream lip
25 58c together form a slotted feed gap 70 through which adhesive 72 flows onto the top of the lower layer 68 as the web travels in left-to-right direction, as illustrated in FIG. 6. This forms a top adhesive layer 74 of the multilayer adhesive product. Again, for ease of illustration, the top layer 74 is shown as a darker-colored material, but this may not necessarily be the case in actual production; for instance,
30 various colors or tags such as ultra-violet fluorescent dye may be utilized to facilitate measurement of individual layer thicknesses.

Coating of viscous adhesives at these web speed rates can involve a number

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of problems. For example, recirculations in the flow of either the bottom or top adhesive layers can result in certain defects in the final multilayer product. Such recirculations may occur if the separation point of either liquid adhesive with respect to the die lips 58 occurs at an inappropriate location. In addition, extreme pressure gradient can result in the upstream leakage of liquid out of the coating gap area, again causing defects in the end product due to nonuniform adhesive layer thicknesses, etc. Moreover, these and other maleffects result in the diffusion of one layer in the other, since they are being coated simultaneously in the liquid state. Such diffusing jeopardizes the integrity and performance of the resulting product.

Thus, it has been found, with respect to the multilayer die coating described herein, that it is very important to control the pressure gradients of the adhesives under each lip. In particular, the top layer should separate from the middle lip at the downstream corner of this lip. In order to achieve such coating control, it will be noted from FIG. 6 that the lips 58 of each die section are stepped or spaced away from the web 52 in the downstream direction. This allows the lips to generate the appropriate pressure gradient and to ensure smooth flow of the adhesive and uniform layer thicknesses. The adjustment of a number of run parameters are necessary in order to achieve this goal. For example, the coating gaps at lip 58b and 58c should be approximately in the range of one to three times the compounded wet film thicknesses of the layers being fed from upstream of said lip. Under the upstream lip 58a, the net flow rate is necessarily zero, and a turn-around flow is the only possibility. Thus, the coating gap under this lip is solely set in order to avoid leakage of the liquid out from the coating gap in the upstream direction. Moreover, the upstream step, defined as dimension A in FIG. 6, and the downstream step, defined as dimension B, may range anywhere from zero to four mils (.0 inches to .004 inches). The feed gaps (defined as dimensions C and D in FIG. 6) can also be adjusted anywhere between one and fifteen mils (0.001 inches to 0.015 inches), preferably not to exceed five times the wet film thickness of their correspondent layers. In addition, the length of the lips 58 in the direction of web travel play an important role in achieving the proper pressure gradient. Thus, the upstream lip 58a should be approximately two millimeters in length, or more, as necessary to seal the head as noted above. The downstream lip 58c and middle lip 58b may fall within

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the range of .1-3 mm in length.

It will be recognized that one of ordinary skill in the art can adjust these various parameters in order to achieve the proper fluid dynamics for uniform layer coating. Of course, persons of more than ordinary skill can adjust the die and run parameters more precisely in order to achieve good results. However, such persons are not always readily available in production settings. Therefore, it is advantageous to provide a die geometry which will increase the size of the window of successful multilayer coating operation. This can be achieved by certain adjustments in the orientation of the die lips.

Thus, FIG. 7 illustrates the die 50 of FIG. 6 rotated slightly in the clockwise direction representing an "angle of attack α ". For consistent reference, the angle of attack (α) shown in FIG. 7 represents a negative angle of attack, or a "converging" orientation of the downstream lip 58c with respect to the web 52. This converging lip orientation provides a negative pressure gradient (in the direction of web travel), along the downstream lip 58c, which is beneficial in preventing a coating defect well known as "ribbing," a pattern of regular striation in the sense of the web travel in the film. The fact that the middle and the upstream lips 58a and 58c also achieve a convergent orientation is not particularly beneficial. Although the angle of attack of the die can be varied widely in order to achieve these advantages, it has been found that angles in the rate of 0° to -5° are appropriate.

An even further successful operating window can be achieved with additional lip modifications. Shown in FIG. 8 is a variation of the lip configuration of FIG. 7, illustrating "beveled" lips. In this configuration, the downstream lip 58c is angled or beveled to as to have a converging profile, similar to that shown in FIG. 7. However, the middle lip 58b is positioned so as to be flat or parallel with respect to the web 52. The upstream lip 58a, on the other hand, is beveled so that it is diverging from the web 52 in the downstream direction. This configuration, again, provides the appropriate pressure gradient under the individual lips to avoid recirculations and upstream leakage. Moreover, if perturbations in the coating conditions occur (such as, for example, due to roll run-out, foreign objects on the web, variations in ambient pressure, etc.), the converging configuration of the upstream lip 58a shown in FIG. 8 will produce a dampening effect on flow

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conditions so that defects in the coating layers do not occur. In this manner, the multilayer coating bead acts as a nonlinear spring to dampen out such unwanted events in order to return to steady state. The die 50 can then be adjusted in accordance with standard angle of attack variations to achieve favorable coating conditions. Because the lips 58 are pre-disposed or beveled in a favorable orientation, the adjustment of angle of attack, as well as coating gap, need not be so precise. Thus, persons of ordinary skill or even less skill can successfully achieve good coating results.

It will be understood, however, by those of ordinary skill in the multilayer coating art, that multilayer die coating can be achieved in a variety of ways similar to those described above, or in other ways, and with appropriate adjustments of the various parameters for coating.

CONVERTING EXPERIMENT

To assess the converting characteristics of pressure-sensitive adhesive constructions of the present invention, a converting trial was performed.

A dual die apparatus was used to hot melt coat the adhesive formulations of Table I to form the adhesive constructions disclosed in Table III. The Lower Adhesive Layer, as designated in Table III, was coated on a 15" wide release liner consisting of 40 lb. silicone coated backing paper. The Upper Adhesive Layer, as disclosed in Table III, was then coated on the Lower Adhesive Layer substantially simultaneously by the dual die apparatus. A 60# High Gloss white facestock was then laminated to the Upper Adhesive Layer to form the final pressure-sensitive adhesive construction. The formulations coated and the coat weight of the respective adhesive layers was varied in accordance with the data disclosed in Table III, and the Examples numbers in Table V, below, denote the equivalent construction from Table III.

As a control, pressure-sensitive adhesive constructions, similar to conventional pressure-sensitive adhesive constructions in having one adhesive composition with its characteristic glass transition temperature, were formed with Formulations 1, 6, and 7 at coat weights of 20 g/m², using the same facestock and release liner. These laminated controls were equivalent to Examples 5, 15, and 18 of Table III, respectively.

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5 The control and multilayer pressure-sensitive adhesive constructions were then converted through a Mark Andy Model 1420 press utilizing an X-die (4 labels across, 15/16" label width, 3.42" label length, 3/32" radius rounded label corner, with a 1/16" cross direction matrix and a 7/64" machine direction matrix), and a V-die (1 label across, 15/16" label width, 14" label length, 1/16" cross direction matrix, 1/2" machine direction matrix). After the die cutting operation, the matrix surrounding the labels was peeled away to leave rows of four rectangular labels adhered to the release liner in the case of the X-die conversion, and one label across in the case of the V-die conversion.

10 The efficacy of the converting process was monitored by the press operator, who observed the die cutting and matrix stripping operations. The existence of "hangers," a term used to describe an adverse converting condition where pieces of the matrix are left on the release liner, were noted as a function of the press speed. Faster press speeds and an absence of hangers is indicative of a construction which
15 converts better. In addition, label predisensing (another adverse converting condition) was also noted, although this condition is less dependent on converting speed than are the formation of flags and hangers.

The data from the converting trials are disclosed in Table V. The reported speed in Ft/Min is the maximum converting press speed that could be obtained
20 without experiencing hangers. This data demonstrates that the multilayer construction of the present invention show improved converting performance in relation to conventional pressure-sensitive adhesive constructions using similar adhesive compositions. For instance, when converted as above Example 3 showed no hangers at converting speeds of up to 800 feet per minute. In contrast, for the
25 control, Example 5, hangers were noted at an approximate press speed of 650 feet per minute, and press speed had to be reduced to 600 feet per minute. Similar results are shown for Example 12 and for Example 16 in comparison to the control, Example 18. Furthermore, although not disclosed in Table V, multilayer adhesive constructions made in accordance with the present invention and formed of acrylic
30 emulsion systems exhibited improved convertability. Thus, the multilayer constructions of the present invention converted at higher speeds with less adverse effects than did the conventional pressure-sensitive adhesive constructions.

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In a separate converting trial, the effects of increasing coat weight on converting performance were investigated. Formulation 3 from Table I was hot melt coated as a single layer on a 40# silicone coated release layer. A 60# High Gloss white facestock was then applied. Three different coat weights, of 15 g/m², 20 g/m², and 25 g/m² were applied to form three constructions. These constructions were converted through a Mark Andy press, as described above. It was observed that the 15 g/m² coat weight construction converted at 700 feet/min, the 20 g/m² coat weight construction at 650 feet/min, and the 25 g/m² coat weight construction at 315 feet/min. Thus, for the single-layer construction, rubber-based PSA control used in these tests, converting speed was seen to decrease dramatically at coat weights over 25 g/m².

It will be appreciated that certain variations of the present invention may suggest themselves to those skilled in the art. The foregoing detailed description is to be clearly understood as given by way of illustration, the spirit and scope of this invention being limited solely by the appended claims.

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TABLE V.

Adhesive Construction From Table III	Convertibility Using X-Die Ft/Min	Convertibility Using V-Die Ft/Min
5 Ex. 3	800	
Ex. 5	600	
Ex. 11	pred, 800	pred, 750
Ex. 12	pred, pred	900, 900
Ex. 13	890, 885	890, 890
10 Ex. 14	890, 780	740, 740
Ex. 15	pred, pred	890, 900
Ex. 16	pred, pred	890, 890
Ex. 17	870, 800	890, 880
Ex. 18	800, 650	750, 750
15 Ex. 19	850, 550	720, 780
Ex. 22	pred, pred	910, 950
Ex. 23	800, 650	750, 750

WHAT IS CLAIMED:

1. An adhesive construction, comprising:
a facestock;
a first adhesive layer on a first side of the facestock having a first
5 glass transition temperature;
a second adhesive layer on the construction on the first side of the
facestock, the second adhesive layer having a second glass transition
temperature;
wherein the first glass transition temperature is different from the
10 second glass transition temperature.
2. The adhesive construction of Claim 1, wherein the first adhesive layer
comprises a first adhesive composition having at least a first polymeric component,
and the second adhesive layer comprises a second adhesive composition having a
15 second polymeric component, the first adhesive layer being in contact with the
facestock and the second adhesive layer being in contact with the first adhesive
layer, and a release surface on the second adhesive layer.
3. The adhesive construction of Claims 1 or 2, wherein the first glass
transition temperature is within the range of from about 10°C to about 50°C higher
than the second glass transition temperature.
- 20 4. The adhesive construction of any of Claims 1-3, wherein the first
glass transition temperature is within the range of from about 15°C to about 35°C
higher than the second glass transition temperature.
5. The adhesive construction of any of Claims 1-4, wherein at least one
of the first and second polymeric components comprises block copolymers of
25 styrene-butadiene-styrene, styrene-isoprene-styrene, styrene-butadiene, styrene-
isoprene, multibranched styrene-butadiene, multibranched styrene-isoprene, or an
acrylic based polymer.
6. The adhesive construction of any of Claims 1-5, further comprising
at least a first tackifier additive in the first adhesive composition and a second
30 tackifier additive in the second adhesive composition, wherein the tackifier additives
are present in a concentration by weight in the respective first and second adhesive
compositions of from about 40% to about 90%.

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7. The pressure-sensitive adhesive construction of any of Claims 1-6, further comprising a plasticizer present in at least the first or second adhesive composition in a concentration by weight of about 1% to about 30%.

5 8. The pressure-sensitive adhesive construction of Claim 7, wherein at least some of said tackifiers and plasticizers may migrate to an adjacent adhesive layer, and the ratio by weight of the first polymeric component to the sum of the migratory tackifier additives and plasticizers in the first adhesive composition, when divided by the ratio by weight of the second polymeric component to the sum of the migratory tackifier additives and plasticizers in the second adhesive composition is
10 between about 0.8 and about 1.2.

9. The adhesive construction of Claim 1, wherein one or more additional adhesive layers are positioned between the first adhesive layer and the second adhesive layer.

10. A pressure-sensitive adhesive construction, comprising:
15 a facestock;
a first layer on the facestock, the first layer comprising a first polymeric composition with a first glass transition temperature, a first storage modulus, and a first tangent delta;
a second layer on the first layer, the second layer having a second
20 polymeric composition with a second glass transition temperature, wherein the second polymeric composition is also a pressure-sensitive adhesive.

11. The pressure-sensitive adhesive construction of Claim 10, where the first storage modulus at a frequency of 10^4 radians per second at 20°C is greater than about 3×10^8 dynes/cm².

25 12. The pressure-sensitive adhesive construction of Claim 10 or 11, where the first tangent delta at 20°C and 10^2 radians per second is greater than 0.5.

13. A method of producing a multilayer pressure-sensitive adhesive construction with improved convertibility, comprising the steps of:
providing a first surface;
30 applying a first pressure-sensitive adhesive layer to the first surface, wherein the first pressure-sensitive adhesive layer includes a first adhesive composition having a first glass transition temperature;

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applying a second pressure-sensitive adhesive layer to the first pressure-sensitive adhesive layer, wherein the second pressure-sensitive adhesive layer includes a second adhesive composition having a second glass transition temperature which differs from the first glass transition temperature; and

applying a second surface to the second pressure-sensitive adhesive layer.

14. A method of improving the convertibility and aging of a multilayer pressure-sensitive adhesive construction, comprising the steps of:

providing a first elastomeric polymer composition, wherein the first elastomeric polymer composition includes at least a first polymer component and at least a first migratory organic additive, such that the ratio by weight of the first polymer component to the first migratory organic additive defines a first value;

providing a second elastomeric polymer composition, wherein the second elastomeric polymer composition includes at least a second polymer component and at least a second migratory organic additive, such that the ratio by weight of the second polymer component to the second migratory organic additive defines a second value;

wherein the first value is approximately equal to the second value;
applying the first elastomeric polymer composition having the first value to a first surface to form a first adhesive layer;

applying the second elastomeric polymer composition having the second value to the first adhesive layer to form a second adhesive layer; and

applying a second surface to the second adhesive layer.

15. The method of Claim 13, wherein the second surface is a facestock and the second glass transition temperature is greater than the first glass transition temperature by from about 10° to about 50°C.

16. The method of any of Claims 13-15, wherein the first and second adhesive layers are applied substantially simultaneously by a dual die.

17. The method of any of Claims 13, 14, or 16, wherein the first surface is a release surface and the second surface is a facestock.

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18. The method of any of Claims 13, 14, or 16, wherein the first surface is a facestock and the second surface is a release surface.

19. The method of Claim 14, wherein the first elastomeric polymer composition has a first glass transition temperature, the second elastomeric polymer composition has a second glass transition temperature, the second surface is a facestock, and the second glass transition temperature exceeds the first glass transition temperature by from about 10°C to about 50°C.

20. A method of making an adhesive construction with improved convertibility, comprising the steps of:

10 providing a first surface;

applying a first adhesive layer to the first surface, the first adhesive layer having a first adhesive composition with first glass transition temperature;

15 applying a second adhesive layer to a second surface, the second adhesive layer having a second adhesive composition with a second glass transition temperature which differs from the first glass transition temperature by from about 10°C to about 50°C; and

20 laminating the first surface and first adhesive layer to the second surface and second adhesive layer, such that the first adhesive layer contacts the second adhesive layer, to form the adhesive construction.

1/5

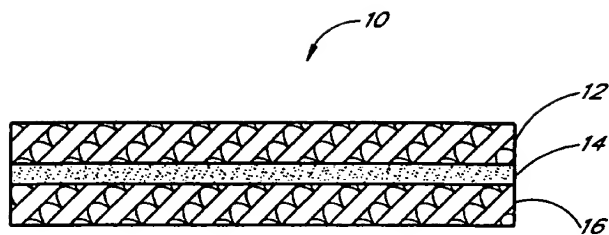


FIG. 1

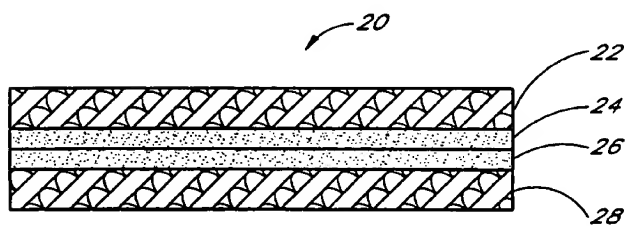
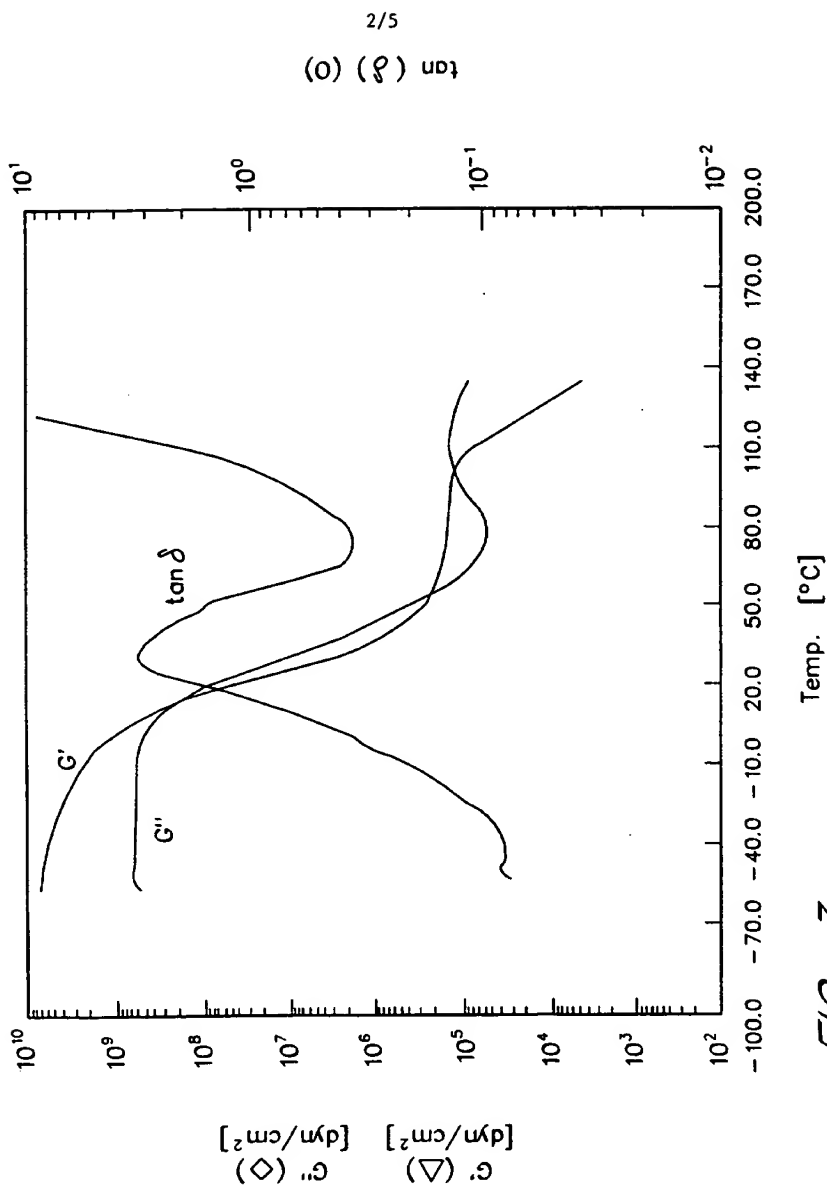


FIG. 2



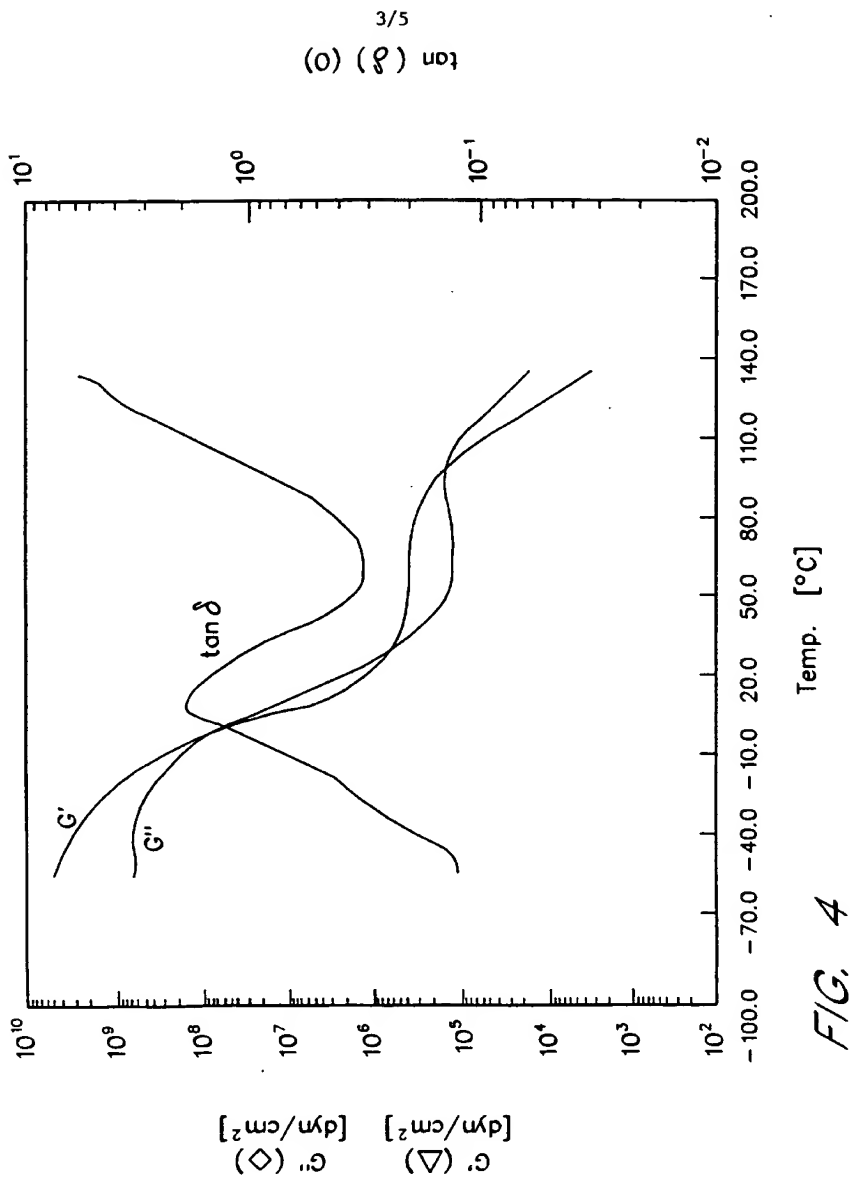
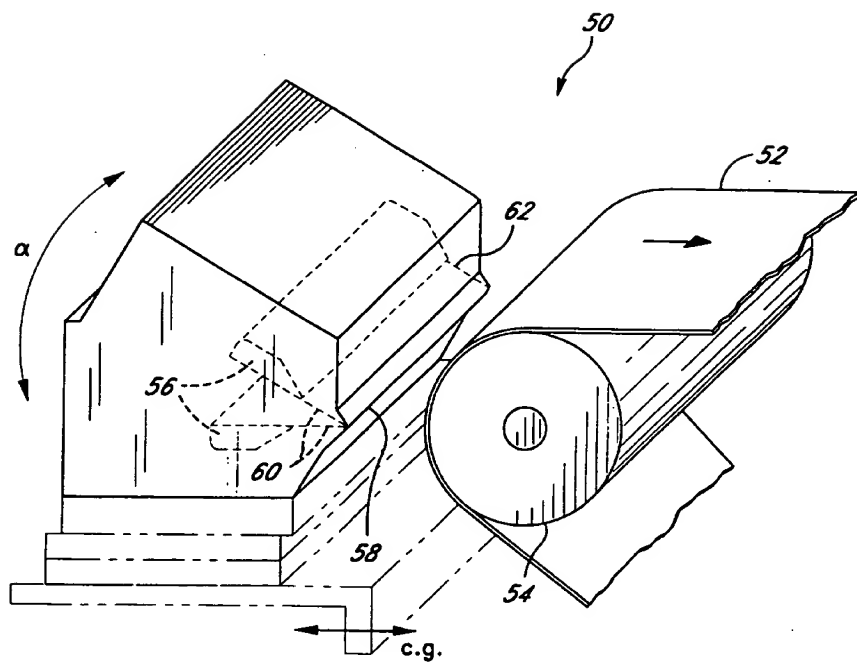
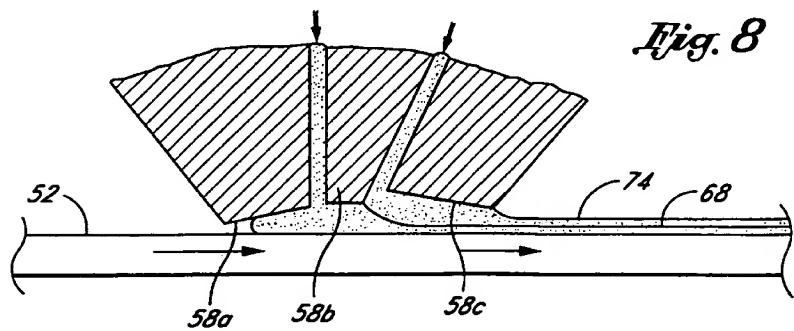
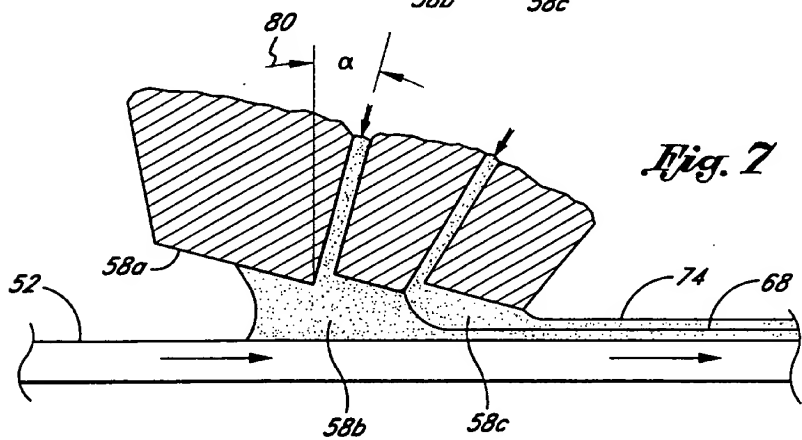
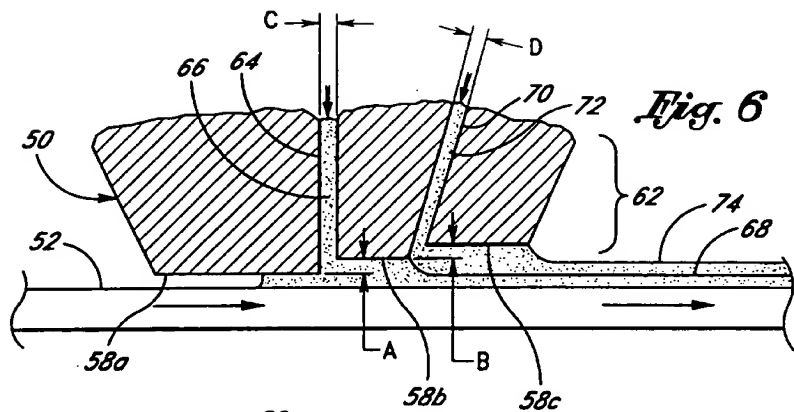


Fig. 5





INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : B32B 7/10, C09J 7/02		A3	(11) International Publication Number: WO 96/08367
			(43) International Publication Date: 21 March 1996 (21.03.96)
(21) International Application Number: PCT/US95/11704		Pasadena, CA 91103 (US). SARTOR, Luigi [IT/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US). SASAKI, Yukihiko [US/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US). DE KONING, Henk [NL/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US).	
(22) International Filing Date: 15 September 1995 (15.09.95)		(74) Agent: SIMPSON, Andrew, H.; Knobbe, Martens, Olson & Bear, Suite 1600, 620 Newport Center Drive, Newport Beach, CA 92660 (US).	
(30) Priority Data: 08/307,944 16 September 1994 (16.09.94) US 08/481,124 7 June 1995 (07.06.95) US		(81) Designated States: AM, AT, AU, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TT, UA, UG, US, UZ, VN, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG), ARIPO patent (KE, MW, SD, SZ, UG).	
(60) Parent Applications or Grants (63) Related by Continuation US 08/307,944 (CIP) Filed on 16 September 1994 (16.09.94) US 08/481,124 (CIP) Filed on 7 June 1995 (07.06.95)		Published <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>	
(71) Applicant (for all designated States except US): AVERY DENNISON CORPORATION [US/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US).		(88) Date of publication of the international search report: 23 May 1996 (23.05.96)	
(72) Inventors; and (75) Inventors/Applicants (for US only): ERCILLO, Jesse, C. [US/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US). MANN, Roger, H. [US/US]; 150 N. Orange Grove Boulevard, Pasadena, CA 91103 (US). JOSEPHY, Karl [US/US]; 150 N. Orange Grove Boulevard,			
(54) Title: MULTILAYER PRESSURE-SENSITIVE ADHESIVE CONSTRUCTION			
(57) Abstract			
<p>The present invention is directed toward a multilayer, pressure-sensitive adhesive construction wherein one layer is formed of a first adhesive composition having a first glass transition temperature, and at least a second layer is formed of a second adhesive composition having a second glass transition temperature. The first glass transition temperature differs from the second glass transition temperature.</p>			

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INTERNATIONAL SEARCH REPORT

Inter. Appl. No.
PCT/US 95/11704

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 B32B7/10 C09J7/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B32B C09J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DE,A,15 94 046 (W. JACKSTÄDT & CO) 21 August 1969 see claims 1,4 ---	1,3-5
X	WO,A,93 02855 (AVERY DENNISON CORP) 18 February 1993 see page 12, line 7 - line 12; claims 1,3 ---	1
X	DE,A,40 16 808 (LOHMANN GMBH & CO KG) 1 August 1991 see claim 1 ---	1
X	US,A,4 260 659 (GOBRAN RAMSIS) 7 April 1981 see claims 1,4,6; table I --- -/-	1,9

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

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- * "A" document member of the same patent family

Date of the actual completion of the international search

3 April 1996

Date of mailing of the international search report

04.04.96

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Authorized officer

Schueler, D

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 95/11704

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	GB,A,2 019 248 (MINNESOTA MINING & MFG) 31 October 1979 see claims 1,10; examples 1,3 ---	1,9
X	EP,A,0 262 271 (LEE PHARMACEUTICALS ;ACUTEK ADHESIVE SPEC INC (US)) 6 April 1988 see example 1 ---	20
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X	EP,A,0 157 566 (PROTECTIVE TREATMENTS) 9 October 1985 see page 8, line 20 - page 9, line 8; claims ---	1
A	WO,A,92 13924 (MINNESOTA MINING & MFG) 20 August 1992 see claim 1 ---	1
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X	PATENT ABSTRACTS OF JAPAN vol. 010 no. 103 (C-340) ,18 April 1986 & JP,A,60 233176 (DAIMATSU KAGAKU KOGYO KK) 19 November 1985, see abstract ---	1,3,5,13
X	EP,A,0 092 999 (SQUIBB & SONS INC) 2 November 1983 see page 10, line 1 - page 11, line 15; claims 1,8,21 ---	1,13,14
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/US95/11704

Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

1. Claims : 1-13,20 Multilayer composition and construction characterized by
glass temperature
2. Claims : 14-19 Method characterized by migratory additives

1. ☒ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.☒ No protest accompanied the payment of additional search fees.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 95/11704

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International Application No
PCT/US 95/11704

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